Materials-structure determination of noncrystalline solids: A hybrid approach

The design of complex materials is one of the fundamental problems in materials theory. The structure of present day materials is no longer limited to crystalline symmetry, but ranges from amorphous and quasiperiodic ordering to a complete/partially disordered state. While the rapid development of *ab initio* molecular dynamics (AIMD) methods, such as Car-Parrinello and density-functional-based approaches, in the past decades brought a major breakthrough in modeling complex materials taking into account the quantum effects accurately, these are nonetheless computationally very expensive for large-scale modeling of non-periodic solids. Despite their overwhelming success, realistic modeling of amorphous materials, involving several thousand atoms using density functional theory (DFT), from scratch is still not feasible (except in few circumstances) even though these methods have the advantage of being highly reliable and accurate.

A very different approach to the materials-structure determination problem for amorphous solids is to address the problem from a hybrid point of view. Since an ideal model of a material should satisfy all available experimental information, one can pose the problem as designing a model satisfying experimental data. However, the information available from experiments is often not sufficient to construct a model configuration, exhibiting *macroscopically* unique physical properties. The experimentally constrained molecular relaxation, or ECMR, developed by Biswas and his collaborators a decade ago particularly addressed this aspect of the problem by adding further information of the materials in the form of topological and chemical constraints for modeling amorphous materials.

ECMR is a classical example of a hybrid approach where one employs experimental information along with a total-energy functional in order to construct a set of structural solutions (“models”) that simultaneously satisfy experimental data and are “thermodynamically” stable to produce a correct physical solution. The hallmark of this approach is that, in the presence of experimental information, an approximate total-energy functional suffices to produce correct structural models. It has been observed that the ECMR approach has the ability to rectify any inadequacies associated with approximate atomic interactions with a judicious choice of experimental data. By systematically reducing the degeneracy of a solution set with additional information, it is possible to identify a very few trusted solutions that can represent the actual structure of a material. The current activity of my group is to generalize this concept and to employ the method in studying multicomponent amorphous systems, for example, metallic and bulk metallic glasses.